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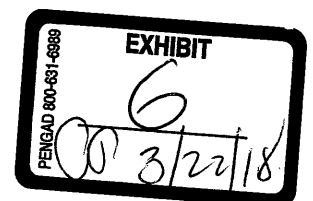
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SYMPOSIUM

Indices for Ranking the Potential for Pesticide Contamination of Groundwater¹

P. S. C. RAO, A. G. HORNSBY, AND R. E. JESSUP²

ABSTRACT

Reports of groundwater contamination by several pesticides and other organic pollutants have increased in recent years. These incidents have prompted regulatory agencies to reevaluate the registration and the use of pesticides for crop production. For this purpose, methods are needed to screen a large number of pesticides to determine their potential to contaminate groundwater. In this paper, several simple indices useful to screen and to rank pesticides in terms of their potential to leach past the crop root zone and to intrude into groundwater are evaluated. These ranking schemes are based on the relative travel time needed for the pesticides to migrate through the vadose zone and on the relative mass emission from the vadose zone (i.e., loading to groundwater). A total of 41 pesticides were screened using six different schemes. Pesticides with solubilities exceeding 10 mg/L and half-lives larger than 50 days seem to have the highest potential. Of these, a number of nematocides and herbicides used in Florida, including EDB, DBCP, aldicarb, carbofuran, bromacil, terbacil, simazine, and cyanazine, were ranked as having a considerable potential to contaminate groundwater. Careful management and regulation of the use of such pesticides as well as groundwater monitoring is warranted in areas with permeable sandy soils and high recharge to groundwater.

Additional Index Words: sorption, degradation, leaching, vadose zone, groundwater pollution.

INTRODUCTION

Reports of groundwater contamination with a number of pesticides and other organic chemicals in Florida and other states have increased in recent years (Pye et al., 1983). In Table 1, various reports of organic contaminants detected in groundwater are summarized. Nematocides, herbicides, and industrial solvents dominate the list in Table 1. Such reports have brought to the forefront public debate, scientific inquiry, and regulatory considerations of the environmental hazards associated with pesticide use in crop production and with land disposal of hazardous organic wastes.

A number of comprehensive computer simulation models are available for site-specific evaluations of the pesticide behavior in the root zone (e.g., Carsel et al., 1984). Models have also been developed to describe the various environmental processes that influence pesticide dynamics in soils (Rao and Jessup, 1983). Such models are usually data intensive and require

knowledge of a number of soil, environment, crop, and pesticide parameters. In a majority of the cases, such parameters are neither available nor likely to be available in the near future due to the high cost associated with obtaining such data for a large number of soil-crop-pesticide combinations. As an alternative, we must look for simple screening approaches for assessing the relative potential of various pesticides to leach beyond the crop root zone and intrude into groundwater.

Several empirical approaches for screening purposes have been proposed. The Arizona Department of Health Services (1982) devised a numerical rating scheme for developing a list of "priority pesticides" that have the highest potential for contaminating groundwater in Arizona. This scheme was based on assigning numerical values to various criteria, including pesticide chemical properties (solubility and persistence), volume of use, and health effects. A similar scheme is now being considered by the Florida Department of Agriculture and Consumer Services for use in Florida (Van Middlem, 1985; personal communication). Aller et al. (1985) have proposed a numerical rating scheme, called DRASTIC, for evaluating the potential for groundwater contamination at a specific site given its geohydrologic setting. The acronym for their scheme is derived from the seven factors considered in the rating scheme: (i) depth to groundwater, (ii) recharge rate, (iii) aquifer media, (iv) soil media, (v) topography, (vi) impact of the vadose zone, and (vii) conductivity of the aquifer. A combination of weights and ratings are assigned to each of these factors, and a numerical rating, called the DRASTIC index, is calculated for each site or an area. This index is then used to prioritize various areas with respect to their vulnerability for groundwater contamination. It should be noted that the DRASTIC scheme does not explicitly take into account the pesticide properties. A number of other approaches, based on numerical ratings of several factors, have also been proposed and are in use for evaluating the suitability of sites for land disposal of hazardous wastes (Seller and Ganter, 1980; LeGrand, 1983; USEPA, 1983; Gibb et al., 1983; MDNR, 1983).

In this paper we propose a quantitative index for screening purposes. Based on published data for several pesticides, the numerical values for the index were calculated. The pesticide ranking based on this index is compared with the ranking based on the indices proposed by Laskowski et al. (1982) and Jury et al. (1983; 1984 a, b, c).

ENVIRONMENTAL FATE ANALYSIS

A vast number of physical, chemical, and biological processes control pesticide behavior in soils (Rao et al., 1983). These processes are dynamic and inter-related. The rate at which each of the processes proceeds is controlled by various soil and environmental factors. The questions asked most often in assessing the potential for pesticide contamination of ground-

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TABLE 1.—TOXIC ORGANIC CHEMICALS FOUND IN GROUNDWATER COLLECTED IN THE U.S.†

Chemical	State(s)	Concentration range (µg/L)	Chemical	State(s)	Concentration range (µg/L)
Acetone	NJ	3,000	1,1-Dichloroethylene	MA, ME, NJ	70-280
Alachlor	NB	0.04	1,2-Dichloroethylene	MA, NY	91-323
Aldicarb	AZ, CA, FL, ME, MO, NC, NJ, NY, OR, TX, VA, WA, WI	1-50	1,2-Dichloropropene	CA, MD, NY	1-50
Atrazine	IA, NB, WI	0.3-3.0	Dinoseb	NY	1-5
α-BHC	CA	6	Dioxane	MA	2,100
β-BHC	CA	4	Ethylbenzene	NJ	2,000
γ-BHC (Lindane)	CA	22	Ethylendibromide(EDB)	CA, FL, HI, GA, SC	0.05-300
Benzene	CT, NJ, NY	30-330	Isopropylbenzene	NY	290
Bis-(2-ethylhexyl)phthalate	NY	170	Methylene chloride	NJ, NY	47-3,000
Bromacil	FL	300	Oxamyl	NY	5-65
Bromoform	DE	20	Parathion	CA	4-6
Butylbenzylphthalate	NY	38	Simazine	CA	1-2
Carbofuran	NY, WI	1-5	Tetrachloroethylene	CT, NJ, NY	717-1,500
Carbontetrachloride	NJ, NY	235-400	Toluene	NJ	55-5,440
Chloroform	NJ, NY	67-490	1,1,1-Trichloroethane	CT, ME, NJ, NY	965-5,440
Chloromethane	MA	44	1,1,2-Trichloroethane	NY	20
Cyclohexane	NY	540	Trichloroethylene (TCE)	NJ, NY, PA	1,530-27,300
Dibromochloromethane	DE, NY	20-55	Trifluorochloroethane	NY	35-135
Dibromochloropropane (DBCP)	AZ, CA, HI, MD, SC	0.02-137	Vinylchloride	NY	50
Di-(n)-butylphthalate	NY	470	Xylene	NJ, NY	59-300
1,1-Dichloroethane	NJ	7			
1,2-Dichloroethane	NJ	250			

†Adapted from: Brumaster, D. E. (1981) and Cohen et al. (1984).

water are: How long will it take a pesticide to get into groundwater? How much of the applied amount arrives at the water table? What is the pesticide concentration in the groundwater and does it exceed the allowable limits set by the regulatory agency? In answering these questions, we have to evaluate the following factors:

- (1) Travel time and persistence in the crop root zone and in the intermediate vadose zone,
- (2) Pesticide mass emissions from the vadose zone, i.e., loading to groundwater,
- (3) Pesticide concentration in the groundwater, and
- (4) Pesticide persistence and dilution in the saturated zone.

The total amount of a pesticide leaching past the vadose zone and arriving at the water table is defined here as the mass emission. This amount is determined by the total travel time in the vadose zone. The travel time, in turn, determines the time available for pesticide attenuation within each zone via chemical and biological processes (degradation, plant uptake, etc.). For a given mass emission, the actual concentration found in the groundwater is determined by persistence in the saturated zone and dilution that occurs as a result of groundwater flow. A detailed account of the variety of mechanisms contributing to contamination and attenuation of pollutants in aquifers is given by Freeze and Cherry (1979) and Mackay et al. (1985).

In order to evaluate pesticide behavior in soils, a number of pesticide properties, referred to as the chemodynamic parameters is needed. These include: aqueous solubility expressed either on the basis of mole fraction (Xw) or mass per volume (S), melting point (MP), vapor pressure (Vp), Henry's constant (Kh), octanol-water partition coefficient (Kow), sorption coefficient (Koc), and degradation half-life

(t_{1/2}). Values for these parameters are required by USEPA and state regulatory agencies as a part of the pesticide registration process. Published data on these parameters have been compiled and summarized by a number of authors (e.g., Rao and Davidson, 1980; Karickhoff, 1981; Kenaga and Goring, 1980; Lyman et al., 1982). These chemodynamic parameters are inter-related. Empirical and theoretical expressions have been proposed to estimate the values of one parameter using other chemodynamic parameter(s) or more fundamental chemical properties. For example, pesticide sorption coefficient (Koc) can be estimated given the aqueous solubility (Xw or S) and melting point (MP) or octanol-water partition coefficient (Kow). Lyman et al. (1982) present a compendium of data and methods for estimating the chemodynamic parameters for pesticides and other organic pollutants.

In addition to the pesticide chemodynamic parameters, detailed characterization of the pesticide behavior in soils requires the following site-specific information: climatological data including daily records of rainfall, evapotranspiration, temperature, and net radiation; irrigation, crop, and pesticide management practices; soil profile characteristics including depth to groundwater (L), total porosity (P), volumetric soil water contents at field capacity (FC) and permanent wilting point (PWP), soil bulk density (BD), soil organic carbon content (OC), and groundwater net recharge rate (q); and crop parameters such as rooting depth (L₁) and rooting density.

INDICES FOR GROUNDWATER CONTAMINATION POTENTIAL

ATTENUATION FACTOR AND RETARDATION FACTOR

Given detailed information as described above, sophisticated simulation models may be used to predict the pesticide concentration distributions in the

soil profile and the total pesticide losses beyond the root zone throughout the season. The pesticide root zone model, PRZM, developed by the USEPA (Carsel et al., 1984), is an example of such models. Since our intent here is not a detailed prediction, but the development of a simple scheme for ranking the relative potentials of different pesticides to intrude into groundwater, we may be able to utilize much simpler methods. For such a purpose we envision the system of interest in two distinct zones: the crop root zone, and the intermediate vadose zone. Our purpose here will be to calculate the travel times through each of these zones as well as to estimate mass emission from the vadose zone and therefore pesticide loading into groundwater.

Assuming that pesticide degradation follows first-order kinetics, the total amount (M_1) of pesticide leaching past the crop root zone is given by:

$$M_1 = M_o \exp(-tr_1 k_1) \quad (1)$$

where M_o is the amount of pesticide applied at the soil surface, and tr_1 and k_1 are the travel time and the first-order degradation rate coefficient in the root zone, respectively. The pesticide amount leaving the root zone is further degraded as it travels through the intermediate vadose zone. Thus, the amount (M_2) entering the groundwater is equal to:

$$M_2 = M_1 \exp(-tr_2 k_2) \quad (2)$$

where M_2 , tr_2 , and k_2 are as defined for Eq. (1) except that the subscript 2 designates the values for the intermediate vadose zone. Substituting Eq. (1) in Eq. (2) yields:

$$M_2 = M_o \exp - [(tr_1 k_1) + (tr_2 k_2)] \quad (3)$$

From Eq. (3) it is evident that computation of pesticide mass emission from the vadose zone (i.e., M_2) requires the estimates of the travel times (tr_1 , tr_2) and the rate coefficients (k_1 , k_2) in the root zone and the intermediate vadose zone. Water and solute transport processes within the root zone are transient in nature and the travel time (tr_1) is a complex function of rainfall, irrigation, evapotranspiration, and rooting patterns. Thus, it is difficult to obtain a simple estimate of tr_1 . On the other hand, the difference between annual evapotranspiration and rainfall plus irrigation can be used to estimate tr_2 because the flow processes in the intermediate vadose zone are essentially steady. A considerable amount of data exists for estimating pesticide degradation rates in the root zone based on measurements with surface soils. In contrast, there is a paucity of data for estimating degradation rates in the intermediate vadose zone. In the absence of better information, at this point we are then faced with the need to assume that $k_1 = k_2 = k$. The total travel time (tr) required for a pesticide to travel through the root zone and the intermediate vadose zone is the sum of the respective travel times in each of these zones; thus, $tr = (tr_1 + tr_2)$.

With the above simplifying assumptions, Eq. (2) can be restated as:

$$M_2 = M_o \exp(-tr k) \quad (4)$$

Noting that the pesticide half-life, $t_{1/2} = (0.693/k)$, and defining $B = (0.693 tr/t_{1/2})$, Eq. (4) simplifies to:

$$M_2 = M_o \exp(-B) \quad (5)$$

Finally, the fraction of surface-applied pesticide that reaches the groundwater is equal to:

$$AF = (M_2/M_o) = \exp(-B) \quad (6)$$

where AF is defined as the *Attenuation Factor* and serves as an index for pesticide mass emission from the vadose zone. It should be noted that $0 < AF < 1$.

The total travel time (tr) in Eq. (4) can be approximated by:

$$tr = (L RF FC/q) \quad (7)$$

where

$$RF = [1 + (BD OC Koc)/(FC) + (AC Kh/FC)] \quad (8)$$

L is the distance from the soil surface to groundwater, and AC is the air-filled porosity. Note that we have implicitly assumed that the net recharge rate (q) is the same in both the root zone and the intermediate vadose zone. The limitations associated with such an assumption will be discussed in a later section. The term RF in Eqs. (7) and (8) is known as the *Retardation Factor* (see Davidson et al., 1968; Rao and Jessup, 1983) and represents the retardation of pesticide leaching through soils due to sorption and pesticide partitioning between the vapor and liquid phases. We note that, for non-volatile non-adsorbed pesticides, Koc and $Kh = 0$ and $RF = 1$. Thus, with increasing Koc or Kh the numerical value of RF becomes larger. This implies, then, that the travel time (tr) for a pesticide with a large Koc or Kh is greater than that for a pesticide with a small Koc or Kh . Since Koc of non-ionic pesticides is generally found to be inversely related to pesticide aqueous solubility (S), we can expect the more soluble pesticides to leach through the root zone and the intermediate vadose zone more rapidly than less soluble pesticides. This is the basis on which Cohen et al. (1984) have proposed that pesticides with aqueous solubility exceeding 30 mg L⁻¹ pose a greater threat to groundwater.

We note that the B value is inversely proportional to $t_{1/2}$ and directly proportional to tr . Thus, persistent and soluble pesticides (i.e., those with large $t_{1/2}$ and small Koc) would have smaller values of B and larger values of AF , which implies that a larger fraction of the pesticide is likely to arrive at the groundwater. Conversely, pesticides with low aqueous solubility (large Koc) and/or those that degrade readily to nontoxic forms (small $t_{1/2}$) would have a large B value and a small AF value, and the fraction of the applied amount leaching to groundwater is likely to be small. Thus, AF value can be used as a simple index for ranking a number of pesticides.

Pesticides can also be ranked simply on the basis of the relative travel times through the root zone and the intermediate vadose zone. This would be equivalent to ranking pesticides based on the RF values, without regard to mass emissions. This method would provide the most conservative estimate of the potential for groundwater contamination. In the absence of any information on pesticide half-lives either in the root zone or in the intermediate vadose zone, we recommend the use of RF values to rank the pesticides. It should be recognized that, while pesticides with small AF values (or large RF values) would have a lower

potential to contaminate groundwater, these compounds pose a greater threat of contaminating surface water bodies via runoff (Rao et al., 1983).

COMPARISON WITH OTHER INDICES:

Laskowski et al. (1982) proposed an index for evaluating a pesticide's potential to contaminate groundwater. Their index, designated as LEACH, is calculated as:

$$\text{LEACH} = (S \, t_{1/2}) / (V_p \, K_{oc}) \quad (9)$$

where all terms are as defined earlier. We note that the numerical value of LEACH is directly proportional to aqueous solubility (S) and half-life ($t_{1/2}$), and inversely proportional to vapor pressure (V_p) and sorption coefficient (K_{oc}). Thus, water-soluble and persistent pesticides would have larger values of LEACH indicating a considerable potential to contaminate groundwater. Volatile pesticides and those with low solubility or short half-lives would receive a lower ranking. Though the index LEACH derived in this scheme lacks a physical meaning, it is intuitively consistent in incorporating the effects of pesticide properties on leaching.

Laskowski et al. (1982) also proposed an index, VOLAT, for assessing the volatilization potential of pesticides. This index is defined as:

$$\text{VOLAT} = (V_p \, t_{1/2}) / (S \, K_{oc}) \quad (10)$$

Larger volatilization potentials, indicated by large values of VOLAT, are associated with pesticides having large vapor pressures (V_p), large half-lives ($t_{1/2}$), and low sorption coefficients (K_{oc}) or aqueous solubility (S). We must note that pesticides with larger volatilization potentials would have a lower potential for groundwater contamination.

Jury et al. (1983; 1984a, b, c) proposed a scheme for ranking pesticides on the basis of a "benchmark" approach. They showed that the travel time (T_c) by convective flow in soil-water, assuming steady water flow, is:

$$T_c = (TH \, RF \, L) / q \quad (11)$$

where TH is the volumetric soil-water content and other parameters are as defined earlier. We note that, with $TH = FC$, Eq. (11) and Eq. (7) are equivalent. At a given site, the parameters FC , L , and q are the same for all pesticides. Thus, ranking pesticides by the travel time is equivalent to ranking by their retardation factors (i.e., by RF values).

Jury et al. (1983; 1984a, b, c) recognized that, for volatile pesticides, transport via vapor-phase diffusion might be more rapid than convective transport in the soil-water phase. They showed that the time for vapor-phase transport (T_d) is given by:

$$T_d = \left[\frac{L^2 \, P^2 \, FC \, RF}{D_g \, AC^{10/3} \, K_h} \right] \quad (12)$$

where P is total porosity and is equal to ($AC + FC$), D_g is the molecular diffusion coefficient in the vapor-phase, and other parameters are as defined earlier. From Eq. (12) we note that pesticides with large D_g or K_h values would have smaller diffusion times for traveling to the water table. Note that, with decreas-

ing air-filled porosity (AC), T_d values becomes large because a smaller cross-section of the pore space is available for vapor transport and pesticide movement would be mostly in the liquid phase.

COMPARISON OF THE INDICES FOR SOME PESTICIDES

For several pesticides, the groundwater contamination potential indices discussed in the previous section were computed (See Eqs. 6, 8-11). For this purpose the chemodynamic parameters for a total of 41 pesticides commonly used in the U.S. were compiled (Table 2). A broad spectrum of pesticides with solubilities and vapor pressures varying over 8 orders of magnitude are included in Table 1. Many of these pesticides are also used extensively in Florida (e.g., aldicarb, carbarthyl, carbofuran, atrazine).

Two soils were selected for computing the indices. Tavares fine sand (Typic Quartzipsamments) was chosen as a representative of the well-drained coarse textured soils of Florida's central ridge, while the soil in the Florida panhandle were represented by Orangeburg loamy sand (Typic Paleudults). Soil properties needed for computing the various indices are summarized in Table 3. These values were obtained by calculating depth-weighted averages using the data reported by Calhoun et al. (1974) for a 2-m deep soil profile. In both soils, the depth to the water table (L) was assumed to be 10 m and the net groundwater recharge rate (q) was set at 0.38. Both these values are fairly typical for the areas where these soils are found in Florida.

The 41 pesticides were ranked in terms of their potential to contaminate groundwater on the basis of each of the six indices presented earlier. Pesticides were ranked in descending order of their potential to contaminate groundwater. Thus, within each scheme a rank of 1 was assigned to the pesticide with the greatest potential, while a rank of 41 indicates the pesticide (among those we considered) with the least potential to contaminate groundwater. Ranking of the 41 pesticides by the six schemes is compared in Tables 4 and 5. Recall that rankings using travel times (T_c) and by retardation factors (RF) are identical; hence the ranks assigned by both these methods are the same. The ranking based on the attenuation factor (AF) and LEACH index accounts for retardation of pesticide leaching due to sorption and pesticide losses due to degradation. In Tables 4 and 5, the pesticides are listed in ascending order of ranks assigned by the AF scheme. This was done to facilitate comparison of ranks assigned by the other schemes.

Examination of the pesticides receiving the top 10 ranking by the AF scheme indicates that these are the water-soluble pesticides ($S > 10 \, \text{mg L}^{-1}$) with half-lives exceeding 40 days. By comparing the pesticide ranking by AF and LEACH, we note that 15 of the top 20 pesticides are the same on both the lists, although the exact rank for each pesticide may not correspond. Many of these pesticides are also those that have been detected in groundwater (see Table 1). Thus, the ranking schemes AF and LEACH seem to provide a reasonable indication of the potential for groundwater contamination by different pesticides.

The pesticide ranking by the AF scheme is essentially identical for both soils (Tables 4 and 5) ex-

TABLE 2.—CHEMODYNAMIC PROPERTIES FOR SEVERAL PESTICIDES.†

Pesticide	S (mg L ⁻¹)	K _{oc} (m ³ kg ⁻¹)	K _h	V _p (Pa)	t _{1/2} (days)
Alachlor	2.420E+02	1.900E-01	1.300E-06	2.900E-03	7
Aldicarb	9.000E+03	1.000E-02	1.000E-04	1.300E-02	28
Atrazine	3.200E+01	1.600E-01	2.500E-07	4.000E-05	71
Bromacil	8.200E+02	7.200E-02	3.700E-08	3.300E-05	350
Captan	3.300E+00	3.300E-02	4.900E-05	1.300E-03	3
Carbaryl	4.000E+01	2.290E-01	1.400E-03	6.700E-01	22
Carbofuran	3.200E+02	2.800E-02	3.100E-07	2.700E-03	40
Chlordane	1.000E+00	3.800E+01	2.200E-04	1.300E-03	3500
Chlorpyrifos	2.000E+00	6.070E+00	1.800E-04	2.500E-03	63
Cyanazine	1.710E+02	1.680E-01	1.200E-04	2.000E-01	108
2,4-D	9.000E+02	2.000E-02	5.600E-09	5.300E+01	15
DBCP	1.000E+03	7.000E-02	1.700E-02	1.060E+02	180
DDT	3.000E-03	2.400E-02	2.000E-03	2.500E-05	3837
Diazinon	4.000E+01	8.500E-02	5.000E-05	9.700E-05	32
Dieldrin	1.500E-01	1.200E+01	6.700E-04	4.000E+02	868
Disulfoton	2.500E+01	1.600E+00	1.100E-04	2.400E-02	5
Diuron	3.700E+01	3.800E-01	5.400E-08	4.100E-04	328
EDB	3.400E+03	4.400E-02	3.500E-02	1.500E+03	3650
EPTC	3.700E+02	2.800E-01	5.900E-04	4.500E+00	30
Ethoprophos	7.500E+02	1.200E-01	6.000E-06	4.650E-02	50
Fenamiphos	7.000E+02	1.710E-01	2.400E-08	1.330E-04	10
Fonofos	1.300E+01	6.800E-02	2.200E-04	2.800E-02	60
Heptachlor	5.600E-02	2.400E+01	1.450E-01	5.300E-02	2000
Lindane	7.500E+02	1.300E+00	1.300E+04	5.600E-03	266
Linuron	8.100E+01	8.600E-01	2.500E-06	2.000E-03	75
Malathion	1.450E+02	1.800E+00	5.000E-06	5.300E-03	1
Methyl Bromide	1.300E+04	2.200E-02	1.500E+00	5.200E-05	55
Methyl Parathion	5.700E+01	5.100E+00	4.400E-06	1.300E-03	15
Monuron	2.600E+02	1.800E-01	7.600E-09	6.700E-05	166
Napropamide	7.300E+01	3.000E-01	7.900E-07	5.300E-04	70
Oxamyl	2.800E+05	6.000E-03	9.900E-09	3.100E-02	6
Parathion	2.400E+01	1.100E+01	6.100E-06	5.000E-03	18
Pentachlorophenol	2.000E+01	1.429E+01	8.800E-02	1.600E+01	48
Phorate	5.000E+01	6.600E-01	3.100E-04	8.500E-05	82
Picloram	4.200E+02	2.600E-02	1.900E-08	8.200E-05	138
Prometryne	4.800E+01	6.100E-01	5.600E-07	1.300E-04	60
Propachlor	6.100E+02	4.200E-01	4.400E-06	3.100E-02	7
Simazine	5.000E+00	1.400E-01	3.400E-08	8.100E-07	75
Terbacil	7.100E+02	4.600E-02	8.200E-09	6.500E-05	50
Triallate	4.000E+00	3.600E+00	7.900E-04	1.600E-02	100
Trifluralin	3.000E-01	7.300E+00	6.700E-03	1.400E-02	132

†Adapted from: Jury et al. (1984 b).

cept for one difference. Methyl bromide was ranked 19 in the Tavares soil, while in the Orangeburg soil it received a rank of 7. Since the half-life value for methyl bromide was assumed to be the same in both soils, a higher rank (i.e., a smaller AF value) in the Tavares soil is the consequence of a larger retardation factor (RF = 6.21) resulting from a higher air content (AC = 0.285) compared to the Orangeburg soil (AC = 0.094; RF = 1.47). Since methyl bromide is highly water-soluble ($S = 13 \text{ g L}^{-1}$), retardation of leaching in water due to sorption on soil is negligibly small ($K_{oc} = 0.022$). However, the RF is large because of highly volatile methyl bromide partitioning between

TABLE 3.—SOIL PROPERTIES* USED IN RANKING THE PESTICIDES.

Soil property	Units	Tavares	Orangeburg
Porosity (P)	m ³ m ⁻³	0.367	0.397
Field Capacity (FC)	m ³ m ⁻³	0.082	0.303
Air Content (AC)	m ³ m ⁻³	0.285	0.094
Bulk Density (BD)	kg m ⁻³	1,550	1,610
Depth to Groundwater (L)	m	10.0	10.0
Net Recharge Rate (q)	m yr ⁻¹	0.38	0.38
Organic Carbon Content (OC)	—	0.0009	0.0031

*These values are depth-weighted averages for a 2-m soil profile. Based on the data of Calhoun et al. (1974).

the soil-air and the soil-water phases. It should be noted that this effect is noticeable only for methyl bromide because its K_h value ($K_h = 1.5$) is about 100-fold larger than the next most volatile compound (heptachlor).

Comparing pesticide ranking by AF and RF (or Tc) allows for an evaluation of the need to consider pesticide losses by degradation. Pesticides such as oxamyl, 2,4-D, and captan receive higher ranking by the RF scheme, but because of their short half-lives these same chemicals are ranked much lower by the AF scheme. Attenuation of the pesticide by degradation in transit to the groundwater decreases the potential threat; hence, a lower ranking.

An aspect not covered in the foregoing discussion is the consideration of ranking metabolites along with their respective parent compounds. This is especially important for pesticides and other organic compounds that degrade to produce toxic metabolites, which either have longer half-lives or have greater aqueous solubilities than their parent compounds. As an example, consider the case of fenamiphos, a nematicide used in citrus culture. Because of its large K_{oc} and short half-life, fenamiphos received a rank of 29 among the 41 pesticides we evaluated. However, its oxidative metabolites (fenamiphos sulfoxide and fenamiphos sulfone) have much smaller sorption coefficients and

TABLE 4.—COMPARISON OF THE RANKS ASSIGNED TO 41 PESTICIDES IN TAVARES FINE SAND USING SIX DIFFERENT SCHEMES.

Pesticide	Ranking scheme used					
	RM	AT	T _c	T _d	LEACH	VOLAT
EDB	8	1	8	2	25	40
Bromacil	10	2	10	33	2	8
Picloram	4	3	4	34	3	17
DBCP	11	4	11	3	28	39
Diuron	24	5	24	37	13	23
Monuron	18	6	18	41	6	10
Terbacil	7	7	7	39	4	6
Cyanazine	16	8	16	12	21	31
Fonofos	9	9	9	8	22	35
Carbofuran	5	10	5	25	11	24
Aldicarb	2	11	2	9	8	22
Simazine	14	12	14	35	7	5
Atrazine	15	13	15	29	9	14
Ethoprophos	13	14	13	19	18	26
Diazinon	12	15	12	14	12	16
Lindane	29	16	29	16	17	18
Napropamide	22	17	22	28	16	19
2,4-D	3	18	3	40	30	37
Methyl Bromide	23	19	23	1	1	1
Phorate	27	20	27	13	14	9
Oxamyl	1	21	1	36	5	7
Chlordane	40	22	40	23	27	29
Prometryne	26	23	26	31	15	11
EPTC	21	24	21	7	31	33
Heptachlor	39	25	39	5	39	38
Linuron	28	26	28	26	19	20
Carbaryl	20	27	20	6	35	34
Dieldrin	37	28	37	18	41	41
Fenamiphos	17	29	17	38	10	2
Captan	6	30	6	11	24	27
Alachlor	19	31	19	24	20	13
Triallate	32	32	32	15	34	28
Trifluralin	35	33	35	10	38	32
DDT	41	34	41	22	37	30
Propachlor	25	35	25	21	23	15
Chlorpyrifos	34	36	34	20	32	25
Pentachlorophenol	38	37	38	4	40	36
Disulfoton	30	38	30	17	36	21
Methyl Parathion	38	39	33	30	26	4
Parathion	36	40	36	32	33	12
Malathion	31	41	31	27	29	3

longer half-lives than the parent compound (Lee et al., 1985). Thus, evaluation of the groundwater contamination threat from using fenamiphos should be based on the more stable toxic metabolites (fenamiphos sulfoxide and fenamiphos sulphone) and not on the parent compound. This would be true for any organic pollutant which is known to produce more soluble and more stable metabolite(s).

As expected, several volatile pesticides (with $K_h > 10^{-4}$) receive a higher ranking by using the time for vapor-phase diffusion (T_d) as the criterion. For example, pesticides such as carbaryl, EPTC, heptachlor, methyl bromide, pentachlorophenol, trifluralin, and triallate are ranked as being in the top 15 by the T_d scheme, while they are ranked among the bottom 15 by the AF scheme. This suggests that volatile pesticides can arrive at the water table faster than expected on the basis of convective water flow alone. However, we will have to account for volatile losses of these pesticides at the soil surface and additional losses due to degradation in the soil. Thus, ranking by diffusion time alone might overestimate the pesticide's potential to contaminate the groundwater, in the same manner as ranking by T_c alone would overestimate the potential for pesticide mass emission.

As indicated earlier, a major uncertainty associated

TABLE 5.—COMPARISON OF THE RANKS ASSIGNED TO 41 PESTICIDES IN ORANGEBURG LOAMY SAND USING SIX DIFFERENT SCHEMES.

Pesticide	Ranking scheme used					
	RM	AT	T _c	T _d	LEACH	VOLAT
EDB	7	1	7	2	25	40
Bromacil	12	2	12	33	2	8
Picloram	4	3	4	34	3	17
DBCP	11	4	11	3	28	39
Diuron	24	5	24	37	13	23
Monuron	19	6	19	41	6	10
Methyl Bromide	9	7	9	1	1	1
Cyanazine	17	8	17	11	21	31
Terbacil	8	9	8	39	4	6
Fonofos	10	10	10	8	22	35
Carbofuran	5	11	5	25	11	24
Aldicarb	2	12	2	9	8	22
Simazine	15	13	15	35	7	5
Atrazine	16	14	16	29	9	14
Ethoprophos	14	15	14	19	18	26
Diazinon	13	16	13	14	12	16
Lindane	29	17	29	16	17	18
Napropamide	23	18	23	28	16	19
2,4-D	3	19	3	40	30	37
Phorate	27	20	27	13	14	9
Chlordane	40	21	40	23	27	29
Oxamyl	1	22	1	36	5	7
Prometryne	26	23	26	31	15	11
EPTC	22	24	22	7	31	33
Heptachlor	39	25	39	5	39	38
Linuron	28	26	28	26	19	20
Carbaryl	21	27	21	6	35	34
Dieldrin	37	28	37	18	41	41
Fenamiphos	18	29	18	38	10	2
Captan	6	30	6	12	24	27
Alachlor	20	31	20	24	20	13
Triallate	32	32	32	15	34	28
Trifluralin	35	33	35	10	38	32
DDT	41	34	41	22	37	30
Propachlor	25	35	25	21	23	15
Chlorpyrifos	34	36	34	20	32	25
Pentachlorophenol	38	37	38	4	40	36
Disulfoton	30	38	30	17	36	21
Methyl Parathion	33	39	33	30	26	4
Parathion	36	40	36	32	33	12
Malathion	31	41	31	27	29	3

with the ranking scheme based on AF is that the soil water flux in the crop root zone was assumed to be constant with depth and time. To evaluate the impact of this assumption, we calculated the mass emission at the bottom of the root zone using the simulation model CMIS presented by Nofziger and Hornsby (1985). This model computes soil-water balance and chemical movement in the root zone given daily records of rainfall, irrigation, and evapotranspiration. Thus, this model calculates the travel times in the root zone (tr_1) and on this basis the amount of the pesticide leaching past the 60-cm root zone (i.e., M_1) is estimated. Using this amount as a basis, pesticides can be ranked in terms of their potential to contaminate groundwater. Note that the CMIS model does not account for distribution of volatile pesticides between the gas and water phases nor for the gaseous losses at the soil surface. For 13 of the more mobile pesticides such a ranking was performed. Only the Tavares soil data and rainfall and evapotranspiration records for 1984 from the Citrus Research and Education Center at Lake Alfred, FL were used for this purpose. The pesticide ranking by the CMIS model was then compared with that based on the AF discussed earlier. These results are summarized in Table 6. Eleven out of the 13 pesticides evaluated appear on both the lists. These results suggest that a reasonable ranking of the pesticides can

TABLE 6.—COMPARISON OF PESTICIDE RANKING BY TWO SCHEMES.

Rank	Ranking scheme	
	CMIS*	AF**
1	EDB	EDB
2	Bromacil	Bromacil
3	Picloram	Picloram
4	DBCP	DBCP
5	Monuron	Diuron
6	Aldicarb	Monuron
7	Methyl Bromide	Terbacil
8	Diuron	Cyanazine
9	Terbacil	Fonofos
10	Cyanazine	Carbofuran
11	Fonofos	Aldicarb
12	Carbofuran	Simazine

*Ranking based on the amounts leaching past the 60-cm root zone as determined by the CMIS model of Nofziger and Hornsby (1985).

**Ranking based on the Attenuation Factor (see Eq. 6).

be made on the basis of the AF scheme. We recognize that the specific rank assigned to a particular chemical may vary depending upon the rainfall distribution (i.e., we can expect year-to-year variation in the ranks); the top 10 or 15 pesticides, however, are likely to be the same.

Statistical methods to evaluate the impact of annual variations in rainfall patterns on the potential to leach a given pesticide beyond the root zone have been presented by Jones et al. (1983) and Dean et al. (1984). These techniques involve simulation of the pesticide leaching over several years using historical rainfall records and examining the cumulative frequency distribution of the mass emissions beyond the root zone. It should be evident then that detailed daily records of climatological data as well as site-specific soil characterization data are needed for such an assessment.

ADVANTAGES AND LIMITATIONS OF THE INDICES

The index AF we propose here is not designed to be a predictive tool, but rather a simple method for ranking a number of pesticides in terms of their *relative potential* to intrude into groundwater. We perceive the index to be used by regulatory agencies in a preliminary evaluation of a large number of pesticides to select chemicals for groundwater monitoring programs or to initiate site-specific studies.

The desirability of using readily-available site and pesticide parameters in developing such an index necessitated that a number of simplifying assumptions be made. These include:

- (1) vadose zone properties are independent of depth,
- (2) an average groundwater recharge rate can be computed given local rainfall, irrigation, and evapotranspiration data,
- (3) a Koc value can be estimated for each pesticide, based on the assumption that hydrophobic interactions are dominant, and
- (4) an average $t_{1/2}$ value can be estimated for each pesticide.

For a given site and/or pesticide, one or more of these assumptions may not be valid. For example, strong soil layering and other heterogeneities with

depth would invalidate the first assumption listed above. For ionic pesticides, such as diquat and paraquat, the concept of Koc is not applicable. Because both of these pesticides are cationic and are sorbed on soils by ion-exchange, their leaching is retarded considerably more than that anticipated on the basis of their high aqueous solubility.

Pesticide persistence (i.e., $t_{1/2}$) is influenced by a number of soil and environmental factors such that a single $t_{1/2}$ value may not adequately represent pesticide degradation rates in different soils, in the same soil under different environments, or at different depths in the soil profile at a given site. The numbers and diversity of microbes generally decreases with increasing soil profile depth as a result of decreasing supplies of various substrates and oxygen at the lower depths. This was shown to be the case for aldicarb degradation rates measured in soil samples collected from Florida and Georgia (Ou et al., 1985 a, b). However, whether this finding is also applicable for other pesticides and for soils from other locations must be judged with caution since chemical degradation rates (e.g., via hydrolysis) may increase at greater depths. The microbial populations in the groundwater are primarily oligotrophic and their activity is expected to be considerably lower than that of the eutrophic microbes in the root zone (Wilson and McNabb, 1983). The groundwater environment is likely to be anaerobic, while the root zone is generally aerobic. For these reasons, one might expect the metabolic rates and pathways for degradation of pesticides and other toxic organic pollutants to be different in the saturated zone than in surface soils. Delfino and Miles (1985) present data for several industrial organic compounds and aldicarb nematicide in groundwater samples collected in Florida. Only a limited amount of data for estimating degradation rates (or half-lives) in the intermediate vadose zone is available. Based on literature reviews and laboratory experiments, Rao and Davidson (1980) have concluded that $t_{1/2}$ values may vary by a factor of only two among different surface soils. Recent field studies (Rao et al., 1985; Rao and Wagenet, 1985) support this conclusion and, thus, justify the use of a single $t_{1/2}$ value for each pesticide at a given site to calculate the AF value.

The ranking scheme we proposed here was not based on pesticide concentrations in the groundwater. However, given a mixing depth (Lm) within the saturated zone, and the mass emission from the vadose zone (i.e., M2), the average pesticide concentration expected in the groundwater (C) may be estimated to be:

$$C = [M2/Lm P] \quad (13)$$

where the volumetric water content in the saturated zone is set equal to porosity (P), and M2 is as defined in Eq. (5). The calculated value of C may be compared with drinking water standards or other limits set by the regulatory agencies. It must be recognized, however, that such a computation is dependent on the ability to estimate a "mixing depth". Eq. (13) does not consider the degradation and dilution of the pesticide in the groundwater. Thus, it generally overestimates the possibility that pesticide concentrations in the groundwater might exceed certain acceptable values.

The AF scheme provides a reasonably accurate

ranking for screening a large number of compounds, such that attention can be focused on a much smaller number of pesticides having the greatest potential to contaminate groundwater. Several of the pesticides receiving high ranking are used extensively in Florida. Careful management of these chemicals is warranted, especially in the central ridge of Florida with permeable sandy soils and with high recharge to the Floridan aquifer. Groundwater monitoring programs should also be focused on these pesticides.

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